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### ► To cite this version:

Frédéric Gal, Julie Lions, Zbigniew Pokryszka, Philippe Gombert, Solenne Grellier, et al.. CO<sub>2</sub> leakage in a shallow aquifer - Observed changes in case of small release. 12th International Conference on Greenhouse Gas Control Technologies (GHGT12), Oct 2014, Austin, United States. hal-01062362

**HAL Id: hal-01062362**

**<https://hal-brgm.archives-ouvertes.fr/hal-01062362>**

Submitted on 9 Sep 2014

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## CO<sub>2</sub> leakage in a shallow aquifer – Observed changes in case of small release

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### Abstract

Geological storage of CO<sub>2</sub> in deep saline aquifers is one of the options considered for the mitigation of CO<sub>2</sub> emissions into the atmosphere. A deep geological CO<sub>2</sub> storage is not expected to leak but potential leakage monitoring is required by legislation, as *e.g.* the EU Directive relative to Geological Storage of CO<sub>2</sub>. To ensure that the storage will be permanent and safe for the environment and human health, the legislation require that the CCS operators monitor the injection, the storage complex and if needed the environment to detect any CO<sub>2</sub> leakage and its hazardous effects on the environment (Lions *et al.*, 2014). Various monitoring methods are available for the monitoring of CO<sub>2</sub> storage sites and the environment as listed by the IEA-GHG (IEAGHG, 2012) and the monitoring selection tool (<http://ieaghg.org/ccs-resources/monitoring-selection-tool1>). Geophysical based methods have a greater area of investigation but may suffer from insufficient sensitivities to detect small leakages. At the opposite, geochemical monitoring methods may have insufficient investigation area but may be able to detect more subtle changes even if monitoring in deep environments is not straightforward. Leakage detection is not yet well constrained and research efforts and tests are required to gain confidence into monitoring strategies.

In the framework of the CIPRES project, funded by the French Research Agency, a shallow CO<sub>2</sub> release experiment has been performed in October 2013 in a chalk aquifer from the Paris basin. The Catenoy site has been characterised since March 2013 through several wells set on a straight line oriented along the local flow (see Gombert *et al.*, this conference). Such an experiment is designed to gain confidence in leakage detection in subsurface environments by understanding processes and principles governing seepage occurrence. Contrary to other experiments such as ZERT (Spangler *et al.*, 2010) or CO<sub>2</sub>FieldLab ones (Gal *et al.*, 2013), where gaseous CO<sub>2</sub> was injected directly in the water, the injection was done with water saturated with CO<sub>2</sub> at atmospheric pressure. 10 m<sup>3</sup> of water were pumped from the aquifer, then saturated with 20 kg of food-grade CO<sub>2</sub> and injected during 40 hours between 12 and 25 m depth. Daily monitoring of soil gases and water was performed during injection and post-injection phases (2 weeks duration) in the area previously delimited by a tracer test. The aim is to determine if geochemical methods are accurate enough to allow detecting small release in shallow environments. If successful, such an experiment can help to gain confidence in leakage detection.

As expected, no change was noticed in the unsaturated zone. The shape of gas concentrations distribution at the surface ( $\text{CO}_2$ ,  $\text{O}_2$ ,  $\text{N}_2$ ,  $^4\text{He}$ ,  $^{222}\text{Rn}$ ) observed during the injection is strictly similar to the repartition of gas species observed since March 2013. The main process observed is respiration and no change linked to the injection was highlighted, only seasonal effects.

Slight changes were observed in the saturated zone. The water was collected at 15 m deep excepted for one stratified borehole where water was sampled at 15 and 18 m. The pH of the injected water was lower (mean value:  $5.3 \pm 0.1$ ) than the initial pH of the aquifer (7.1-7.2) due to  $\text{CO}_2$  dissolution. Only two monitoring boreholes set 10 m and 20 m downstream from the injection well may be considered as influenced by the experiment. A probable enrichment in  $\text{HCO}_3^-$  linked to interaction of the  $\text{CO}_2$  saturated water with chalk was noticed, with an enrichment close to +8 to +10% of the initial value. For one borehole the pH value remained nearly stable in relation with pH buffering and in the other borehole a slight decrease was observed (-0.1 to -0.15 pH unit). However this decrease is significant as it is above the instrumental uncertainty of the electrodes. In addition, a slight increase of the electrical conductivity was noticed but it did not exceed +6% compared to baseline data.

Such slight changes in the physico-chemical parameters are related to small variations in dissolved elements. Apart from  $\text{HCO}_3^-$ , the other major ion affected by  $\text{CO}_2$ -water rock-interaction is Ca as the aquifer is mainly composed by calcite. Concentrations increases by +8 to +9% whose amplitude is in agreement with the increase of  $\text{HCO}_3^-$ . Trace elements were also little affected, the main change concerned Sr (+8 to +10% increase).

Modifications occurring during this  $\text{CO}_2$  release experiment have small amplitude as expected but these results highlight that geochemical methods are able to detect small leakages. Consequently, effects were noticed only during a short period of time. It is not possible to determine if all the injected  $\text{CO}_2$  has migrated downwards in the direction of flow or if partial lateral migration has occurred, but post-injection monitoring and boreholes logging 12 days after the stop of injection did not reveal any discrepancy in the water columns. On the other hand, the magnitude of the pH change is consistent with the behaviour of the co-injected tracer (dilution ratio  $\sim 30$ ). In the perspective of getting more information on the remobilisation of trace metal elements, a push-pull test will be performed in 2014 on the basis of the learning of this first experiment.

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